James Acker:
Switching from the ocean to the atmosphere, we now have Zhen Liu, School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, currently at Combustion Research Facility, Sandia National Laboratories, Livermore, CA. He's presenting on the photochemical atmosphere over China.

Thanks for the Giovanni image, Zhen. You can go! (I though Techsters were Ramblin' Wrecks)

Zhen Liu:
Thanks Jim,

The title of my talk today is 'Characterizing the photochemical environment over China'.

In my research on atmospheric chemistry and air quality, I use various satellite data, e.g. OMI NO2 tropospheric columns as indicator of fossil fuel combustion emissions, MODIS AOD 550nm as an indicator of atmospheric aerosol (e.g. PM2.5, PM10) abundance.

GIOVANNI makes it much easier for me to quickly explore those satellite data.

I am going to talk about one study during my Ph.D. to explore the missing source of glyoxal (CHOCHO) over China and its implications. Places where GIOVANNI data is used are annotated.

(1) First some background information. The region of concern is China, a country with the largest population, which is concentrated in the Eastern part of the country.

(1) In the past decades, fast economic growth and development have led to growing anthropogenic emissions, which have been directly seen from space. For example, this is the tropospheric NO2 column from OMI, showing the broad polluted regions in east China Plain, Pearl River Delta, and also Sichuan Basin and Northeast China.

But at the same time biogenic emissions are quite low over these polluted regions -

that is quite interesting and different from the eastern part of U.S.

Satellite observations of aerosol optical depth also show that this is the region with the largest aerosol abundance in the world

The goal of my research is to better understand atmospheric chemistry over such a region, with high anthropogenic emissions, low biogenic emissions, and high aerosol abundance. To achieve this I am going to examine processes regulating the observed chemical concentrations, including emissions, chemistry and transport.

Models at multiple scales are used in my study. I use a 1-D model constrained by observations to scrutinize chemistry; and I use 3-D regional model to examine emissions and regional implications of my findings. And these models are used to analyze data from in situ, aircraft and satellite observations.

I evaluate the 3-D regional chemical transport model performance using satellite observations. First we see that the model does a great job in reproducing NO2 columns observed by OMI.

Then this is a comparison for CO. Similarly, not bad.

Then I did another comparison, for glyoxal (CHOCHO). CHOCHO is a dicarbonyl compound that can be formed during the oxidation of various VOC species. So the model performance can tell us how good the VOC chemistry and emissions are in the model. The model underestimates glyoxal by a lot, compared to the observation.

As a modeler, the first question I asked is, ‘what happened to the satellite?’

James Acker:
VOC is 'volatile organic carbon' ?

Zhen Liu:

Yes Jim, sorry i didn't explain that

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(1) Then I found that actually this underestimation does not only occur in our model, but also in those global models. Then the real scientific question is 'what is the missing source of glyoxal over China in these models?

More background information. Recent years, satellite observations of tropospheric glyoxal vertical columns became available using DOAS technique. This is a map of global distribution of glyoxal from SCIAMACHY during 2003 -2007. Note that Thomas Kurosu does have processed a similar product of tropospheric CHOCHO from OMI, but that has not been validated and used as extensively as SCIAMACHY.

Looking at this map, one would ask – Do we understand the observed abundance of glyoxal with current knowledge of VOC chemistry and emissions? And can we explain why we have these hot spots, for example the hotspots over the tropical ocean, and the mid-latitude hot spot over China? Interestingly, it also has been shown that glyoxal concentrations have been increasing over the years. Obviously, there are many interesting questions to explore.

To date, there have been 3 independent global modeling studies of glyoxal budgets. Although these models are slightly different in terms of glyoxal chemistry, for example, they may or may not consider the aerosol sink of glyoxal, all these models consistently show a significant underestimation of glyoxal over China. Therefore, a missing source of glyxoal over China exists in these models.

The cause may be either chemistry or emissions. However, we can rule out chemistry easily given the fact that with the same chemistry these models are doing fairly well on the same latitude over other regions, such as in the U.S. Therefore, it must be emissions of some glyoxal precursors over China that has issues.

One the right hand side, In order to explore the reasons, we conducted modeling analyses of glyxoal over China using a regional model, REAM. The emission and glyoxal chemistry in our model are consistent with those in the global models. And for now we don't consider the aerosol sink, but we note that this additional sink of glyoxal would only make the magnitude of the missing source even larger. Here we plot the unexplained glyoxal columns by comparing the model result with SCIAMACHY. We chose August because glyoxal has a seasonal maximum in summer and so the satellite can get a large signal. In the following we are going to use this delta glyoxal to represent the missing source.

First, we try to qualitatively examine the spatial distribution of delta glyoxal, to gain some clue of the nature of the missing source. For example, is it biogenic? Or is it from biomass burning? Or else is it anthropogenic? Let's first compare the spatial distribution of delta glyxoal with that of the ASTR fire hot spots, which has been used as an indicator of biomass burning emissions. Obviously there is no consistency here, as the missing source spreads much more broadly than the hot spots. Therefore, biomass burning does not appear to be the missing source we are looking for. And next, this is a map of biogenic isoprene emissions. Interestingly, the missing source only exists over regions with low isoprene emissions, whereas over high isoprene areas, the delta glyoxal is very small. This actually reminds us of the contrast in China and U.S. we see in those global modeling results, since isoprene emissions are generally higher in U.S. than in China, and the model performance in the

Sorry the animation failed here, I have two figures under this population figure

Anyway, spatial distribution suggests the nature of the missing source is most likely anthropogenic

Next, we do some quantitative analyses. Following previous studies of satellite vertical columns, we assume a linearized relationship between glyxoal vertical columns and the precursor emissions, as can be described by this equation, which means that over each grid, glyoxal column from each precursor can be written as a product of the precursor emission and the sensitivity of CHOCHO to this precursor.

But it should be noted that given the 70km resolution of the model, this assumption is only good for those precursors with relatively short lifetimes, such as isoprene, and most of the aromatics; but for long lived precursors, such as acetylene, which has a lifetime of a month, horizontal transport would smear such a relationship. To resolve this, we introduce a background term C0 to represent contribution from long lived precursors. In addition, this

background term also include glyoxal from minor sources, such as ethene and mono-terpenes. In these figures we show breakdown of glyoxal from four groups of precursors.

In these maps i am showing the following information:

As expected, glyoxal from aromatic precursors (ARO1, ARO2) are mostly over to their emission regions, due to their short lifetime,

Isoprene (ISOP) is the most reactive one and contributes the most of glyxoal regardless of its low concentrations, and its contributions are much larger in the south.

But we have to be aware that there are still large portion of glyxoal unexplained in this standard model, so such a dominant role of isoprene might not be real.

C2H2 is creating a regional background of glyoxal due to its long lifetime. And given previous studies suggesting C2H2 emissions to be reasonable over the region, we can rule out C2H2 to be the missing source. Further, since these minor precursors contributions are too small to be important, we also rule out them. Then we can rule out the whole background term. In the following, we fill look for the missing source from these three short-lived precursors.

After ruling out the background term, we can transform the linear equation to translate the unexplained CHOCHO columns into emission amounts of those short lived precursors, including isoprene and aromatics. In other words, we will show how much more emissions of each precursor are needed to explain the missing source.

First, this is the resulting scaling factors for isoprene. Over most of the underestimated regions, we need 6 to 20 times of standard isoprene emissions to explain the delta CHOCHO. This apparently is not plausible because isoprene emissions are constrained by leaf area index from satellite and meteorology parameters, and thus cannot have so large uncertainties.

Furthermore, by comparing the model simulated isoprene with ground observations at 3 sites, we actually see a reasonable agreement. Therefore, isoprene emissions are generally healthy over China and isoprene is not the missing source we are looking for.

the bar plot was supposed to jump out ..

The only suspicious precursors left are aromatics. This figure shows that we typically need to increase aromatics emissions by a factor of 4 -10 to reproduce the observed glyoxal. The scaling factors also vary a lot over different regions. Although these scaling factors are also very large, it is actually consistent with VOC concentration comparison, as shown in this figure, you can see that aromatics concentration in our model are lower by factors of 5 to 50 at the three sites. Given such an agreement, we conclude that the missing source of glyoxal over China is mainly due to the severe underestimation of anthropogenic aromatics emissions.

The bar plot is showing the ratio of the model simulation and the in situ observed precursor concentrations

The bottom line is satellite CHOCHO and in situ measured precursors consistently suggest that underestimated aromatic VOC emissions is the missing source of CHOCHO over China

Using the scaling factor we obtained here, we can get a top-down estimate of aromatic emissions over China. With the top-down emissions, the model can reasonably reproduce the satellite observations of glyoxal. In the meantime, the top-down emissions also bring model to a better agreement with the observed aromatic concentrations, although there are still low biases at Shanghai and PRD, which may be because the model resolution of 70km is insufficient to simulate these urban and suburban environments. It is also possible that errors in other precursors lead to the biased scaling factor of aromatics.

Again, the top is satellite CHOCHO (satellite) and the bottom is CHOCHO precursors (ground measurements), both comparing with our model simulation.

Our top-down estimate suggests more than a factor 5 increase of total aromatic emissions over China, from 2.4 Tg per year to more than 13.4 Tg per year.

This would certainly leads to much higher photochemical oxidants, such as PAN and O3, but I will not talk about those here. Instead, another implication that is more interesting to this session is on secondary organic aerosols (SOA), since aromatics are also major anthropogenic precursors of SOA. Before doing any detailed modeling analyses, I did a rough calculation based on mass balance.

From a recent modeling study by Henze el al, current global aromatics emission at 19 Tg per year gives SOA at 3.5 Tg per year. If we assume aromatic SOA increase proportionally with emissions, then the additional aromatics from China would increase the total aromatic SOA production by more than 50 percent to 5.5 Tg per year, which is significant enough to gain some attention. Meanwhile, we also note that our finding of underestimated aromatics may also explain at least partially why there has been model underestimation of OA previously over China and the outflow region.

PAN = peroxy acetyl nitrate

SOA = secondary organic aerosols

The main conclusion is that the current estimated VOC (aromatics in this case, but also likely for other speices) emissions over China are severely underestimated

satellite observations of VOC oxidants like HCHO and CHOCHO could provide good top-down constraint for these emissions

Hope OMI formaldehyde (HCHO) will be up on GIOVANNI soon

Finally, thanks to my coauthors, my international collegues and my advisor

Thank Jim for having me here and I look forward to more collabaration with GIOVANNI!

Thank you all.

James Acker:
Thank you Zhen. Two questions: what is the main source of isoprene, and where are the aromatics coming from? Industrial byproducts?

Zhen Liu:

isoprene is mainly from biogenic sources, e.g. trees

James Acker:
That's what I had vaguely remembered about isoprene

Zhen Liu:
aromatics are from vehicle emissions, and yes, industrial sources

James Acker:
OK - yes, I think that the U.S. is trying to sell more and more cars in China

Zhen Liu:
yes other sources of isoprene are not comparable

James Acker:
Any other questions?

Zhen Liu:
cleaner cars would help

Thank you Jim.

James Acker:
OK, that's all for our afternoon session. Tomorrow we begin early in the U.S. at 7 AM EDT with five international presenters. Get your coffee on the way in!

Thanks to all.

Amanda Truett:
Thanks. Good stuff! :)

David Mocko:
Interesting session - thanks to the presenters!

Tracy Van Holt:
Thank you!

Joan Labay-Marquez:
Thank you!